

# MgB<sub>2</sub>: synthesis, sound velocity, and dynamics of the vortex phase

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The sound velocity is measured in polycrystalline MgB<sub>2</sub> synthesized from the elements, and the bulk modulus and the Debye temperature are calculated. The conversion of an elastic wave into electromagnetic radiation is investigated in the mixed state. The dynamic parameters of the vortex lattice are estimated.

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## SYNTHESIS AND REAL STRUCTURE

Magnesium diboride powder was obtained by synthesis from the elements Mg (98% pure) and B (99.5% pure) in an argon medium. The powder was sintered in a high-pressure chamber of the anvil-cell type with a lenticular cell. The necessary compression was provided by a hydraulic press with a 6300 kN force. The powder was heated to the required temperature, not by the conventional indirect heating,[1] but by passing a current through the cylindrical samples to be sintered, which gave a more uniform temperature distribution over the volume of the sample. The preliminary compaction of the powder was done by a cold two-sided pressing at pressures of up to 1.3 GPa. The pressed sample was placed in a capsule of graphitic boron nitride, which was mounted in the working channel of the high-pressure chamber. After completion of the sintering cycle the material was cooled under pressure, and then the pressure was reduced to atmospheric. The rate of buildup and relief of the pressure was 0.5 GPa/s. During the heating and cooling stages the temperature was changed at a rate of 100 deg/s. The chosen construction of the cell of the high-pressure chamber provided for hydrostatic compression of the material to be sintered to pressures of up to 4.5 GPa and heating to temperatures of up to 2000 K with holds of up to 300 s.

According to the data of an x-ray analysis the samples contained magnesium diboride MgB<sub>2</sub> and an insignificant amount of manganese oxide MgO.

Microstructural and fractographic studies of the samples were carried out on a Camebax CX-50 setup. The x-ray microspectral analysis established the presence of the characteristic lines of magnesium, boron, and oxygen.

A study of the real structure of the samples was done by the thin-foil and diffraction microanalysis methods with the use of a PÉM-U transmission electron microscope.

The results of the study showed that the samples are characterized by appreciable scatter in the grain sizes: in

addition to very fine particles ( $d \approx 0.1 \mu\text{m}$ ) there are also rather large ones (up to  $30 \mu\text{m}$ ), in agreement with the results of Ref. 2.

Some of the large grains are highly fragmented, apparently as a result of the deformation during the preparation of the sample. It was noticed that those grains had a high density of defects, contained fine microcracks and pores, and had large dislocation pileups (Fig. 1). The data of electron microanalysis also attest to strong fragmentation: most of the reflections were smeared into arcs.

In addition to the fragmented grains, there were also rather large ( $d > 5 \mu\text{m}$ ) low-defect grains having a cubic configuration, which is characteristic of the compound MgO, which crystallizes in a cubic structure.

In certain parts of the structure, in particular, near pores and along the boundaries of individual, clearly faceted grains, the presence of interlayers of an amorphous phase, similar to that observed in Ref. 3, was noted.

## SOUND VELOCITY

As far as we know, no experimental results from measurements of the sound velocity in MgB<sub>2</sub> have been published yet. In imperfect samples the most reliable data on the sound velocity can be obtained only by analysis of the first signal, which has passed through the sample along the shortest path. Interconversion of different modes at inhomogeneities (including at grain boundaries) makes the more distant reflections less suitable for measurements, as does to use of some version of a resonance methods.

The method used in this paper was modified somewhat from that described in Ref. 4. In essence it consists in the measurement, in a fixed frequency interval, of the phase-frequency characteristics of two acoustic lines, consisting of delay lines with a sample between them and delay lines without a sample (Fig. 2). The difference of the phase-

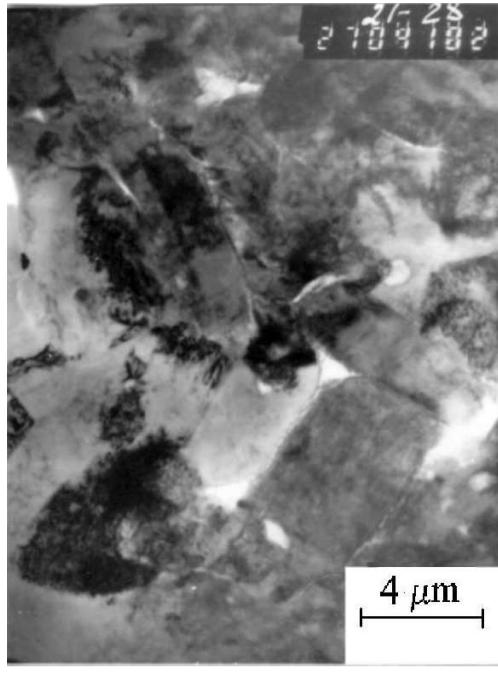


FIG. 1: Typical picture of the real structure of the  $\text{MgB}_2$  sample, demonstrating the substantial inhomogeneity of the structure and the presence of appreciable scatter in the size of the grains.

frequency curves in the absence of interference distortions in the sample is a straight line with a slope determined by the sound velocity that is sought (Fig. 2). For millimeter sample lengths the error of measurement of the absolute value of the frequency at frequencies of  $\sim 55$  MHz lies in the range 0.2–0.3% at a signal-to-noise ratio of  $\sim 3$ .

The velocity values measured at  $T = 77$  K (acoustic path length  $\sim 5$  mm), viz.,  $V_t = 4.83 \times 10^5$  cm/s and  $V_l = 8.2 \times 10^5$  cm/s are only effective values, since defects (pores, microcracks) decrease the sound propagation velocity.[5] The  $\text{MgB}_2$  sample under study had a well-defined geometric shape, making it possible to estimate its porosity ( $\sim 0.13$ ) rather accurately by a simple weighing. Using the approach proposed in Ref. 5, we obtained the corrected values of the velocities:  $V_t = 5.12 \times 10^5$  cm/s and  $V_l = 8.76 \times 10^5$  cm/s. Finally, the limitations imposed in the porous-medium model make this procedure much more uncertain than the error of measurement of the effective values of the velocity. The values obtained are considerably lower than the calculated values,[6] although it may be that the correction introduced is not complete, since it does not make allowance for the presence of microcracks. Starting from the corrected values of the velocities and the x-ray den-

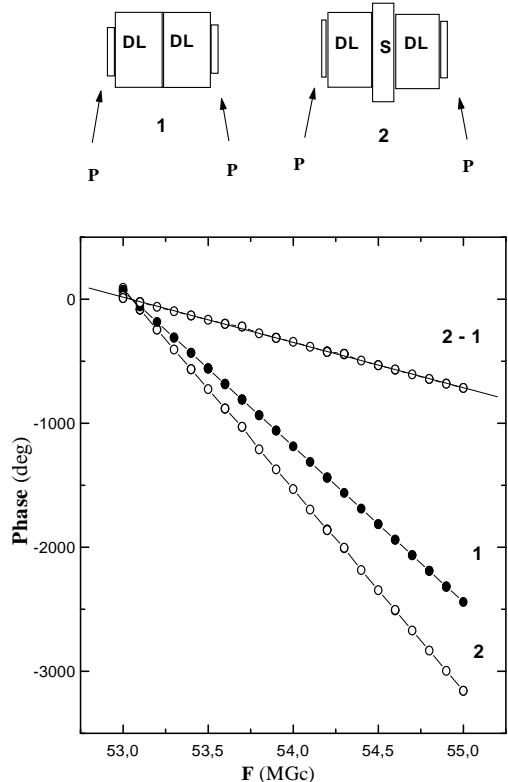


FIG. 2: An example of the determination of the sound velocity  $V$  (transverse sound in  $\text{MgB}_2$ ). The top - the symplified scheme of the experiment . Here P - piezoelectric transducer, DL - delay line, S - the sample. Lower : line 1 - the phase-frequency characteristic (PFC) of two DL only ; line 2 - PFC of two DL and S; line 2 - 1 - the difference between PFC2 and PFC1. Sound velocity is determinated by equation  $V = 360l/A$  ,  $A$  - the slope of the line 2 - 1 ( $365 \cdot 10^{-6}$   $\text{Deg} \cdot \text{sec}$ );  $l$  - the sample length (0.49 cm).

sity, we calculated the bulk modulus ( $B_0 = 110$  GPa) and Debye temperature ( $\Theta_D = 787$  K). The latter value is close to that corresponding to the quantities obtained from heat-capacity measurements ( $\Theta_D = 746$ –800 K; see the literature cited in Ref. 6).

## DYNAMICS OF THE VORTEX PHASE

The critical temperature ( $T_c = 37$  K) and transition width ( $\delta T_c \approx 2$  K) are determined from the measurements of the high-frequency magnetic susceptibility.

To estimate the dynamical parameters of the vortex lattice we have for the first time used the method proposed in Ref. 7, wherein they are extracted from data on the conversion of transverse sound into an electromagnetic field. The sound wave vector  $\mathbf{q}$  was oriented parallel to the external magnetic field  $\mathbf{H}$  and orthogonal to the  $\text{MgB}_2$ -free-space interface. The electromagnetic

radiation was registered by a coil oriented with the plane of its windings also parallel to  $\mathbf{H}$ . The amplitude and phase of the Hall component of the electromagnetic field were measured (the electric field vector  $\mathbf{E}_H$  was orthogonal to the displacement vector  $\mathbf{u}$  in the sound wave). Such experiments had been done before (only the amplitude  $\mathbf{E}_H$  had been recorded) on either low-temperature[8] or high-temperature[9] superconductors, and a simplified theoretical approach to their description is proposed in Ref. 10. However, the relations obtained in Ref. 10 do not admit a correct passage to the limit of the normal state, as will be necessary in order to obtain quantitative information (see below).

A more detailed analysis in the framework of the continuum approximation and the two-fluid model[7] leads to the relation

$$E_H = \frac{1}{c} B \dot{u} \left( \frac{k_L^2 + k_{ns}^2}{q^2 + k_L^2 + k_{ns}^2} \right) \left( \frac{\alpha^*}{\alpha^* + q^2 \frac{B^2}{4\pi} \frac{k_L^2}{q^2 + k_L^2 + k_{ns}^2}} \right), \quad (1)$$

Here  $B$  is the induction in the sample, which is practically equal to the external field  $H$  except in a narrow field region around  $H_{c1}$ . The wave numbers appearing in (1) have the following model dependences on temperature and field:

$$\begin{aligned} k_L^2 &= \lambda^{-2} (1 - t^4) (1 - b), \\ k_{ns}^2 &= 2i\delta^{-2} (1 - (1 - t^4)(1 - b)), \end{aligned}$$

where  $\lambda$  is the low-temperature penetration depth in the Meissner phase,  $\delta$  is the skin penetration depth ( $\delta^{-2} = 2\pi\omega\sigma_n/c^2$ ;  $\sigma_n$  is the conductivity of the normal metal),  $t = T/T_c$ , and  $b = B/B_{c2}$  ( $T_c$  and  $B_{c2}$  are the critical values of the temperature and magnetic field).

The complex quantity  $\alpha^* = i\omega\eta + \alpha_L$  ( $\eta$  is the coefficient of friction and  $\alpha_L$  is the Labusch “spring” parameter) describes the dynamics of the vortex lattice, which vibrates during the propagation of an elastic wave. Bardeen and Stephen[11] gave a simple estimate for  $\eta$ :  $\eta = BB_{c2}\sigma_n/c^2$ . It may be noted that  $\alpha_L \rightarrow 0$  for  $b \rightarrow 1$ .

For  $b = 1$  we have  $k_L = 0$  and  $k_{ns}^2 = 2i\delta^{-2}$ , and Eq. (1) reduces to the known expression for a normal metal in the local limit.[12] In the normal state the phase of  $E_H$  leads  $\dot{u}$  by  $\varphi_0 = \arctan(q^2\delta^2/2)$ , and  $|E_H c/\dot{u}|$  varies linearly with  $H$ , with a slope  $(1 + \tan^2 \varphi_0)^{-0.5}$ . According to published data,[13] for MgB<sub>2</sub> the penetration depth  $\lambda$  lies in the range 800–2000 Å. At the frequencies used here  $q \sim \delta^{-1} \sim 10^3$ , and therefore for  $(1 - b) > 10^{-2}$  and at practically any temperature we have  $k_L \gg q, |k_{ns}|$ , and relation (1) reduces to

$$E_H = \frac{1}{c} B \dot{u} \left( \frac{\alpha^*}{\alpha^* + q^2 \frac{B^2}{4\pi}} \right), \quad (2)$$

which is suitable all the way down to the Meissner state. We emphasize that the factor multiplying  $q^2$  in Eq. (2),

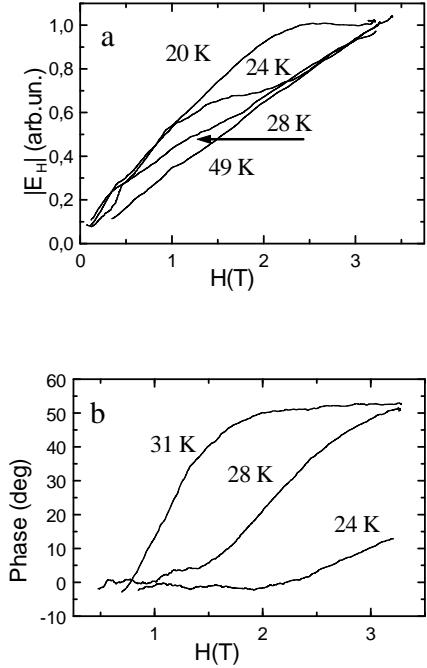


FIG. 3: Field dependence of the phase (a) and amplitude (b) of the Hall component of the radiated electromagnetic field at different temperatures.

unlike the situation in Eq. (1), is not identical to the bending modulus  $C_{44}$  of the vortex lattice, since the latter should soften as  $H_{c2}$  is approached.[14] For  $(1 - b) \ll 1$  ( $\omega\eta \approx 2B_{c2}^2/4\pi\delta^2$ ,  $\alpha_L \approx 0$ ,  $k_{ns}^2 \approx 2i\delta^{-2}$ ) both Eqs. (2) and (1) reduce to the corresponding relation for the normal metal at any value of  $k_L^2$ . This means that even in an ideally homogeneous sample the electric field  $E_H$  changes continuously (without a jump) at the transition of  $H$  through  $H_{c2}$ . As  $H$  is decreased further, the evolution of  $E_H$  depends on the relationship between  $\omega\eta$ ,  $\alpha_L$ , and the elastic modulus  $C_{44} = B^2/4\pi$  of the vortex lattice. Obviously, sooner or later the situation  $|\alpha^*| \gg C_{44}$  will arise, and in that case the phase of  $E_H$  will coincide with the phase of  $\dot{u}$ , and  $|E_H c/\dot{u}|$  will also vary linearly with the field, but now with a unit slope. Thus in the case  $\lambda^{-2} \gg q^2, \delta^{-2}$  relation (2) describes the behavior of  $E_H$  in the entire field interval from  $H_{c2}$  to  $H_{c1}$ .

The measured  $E_H$  curves are presented in Fig. 3. In view of the limited range of magnetic fields accessible in our measurements, informative experiments could be done only at rather high temperatures. We see that the phase of the signal at the transition of  $H$  through  $H_{c2}$  changes by approximately  $\varphi_0 \approx 50^\circ$  (Fig. 3a). The ratio of the slopes ( $\sim 1.6$ ) in the regions of linear variation of  $|E_H|$  is in good agreement with the value that follows

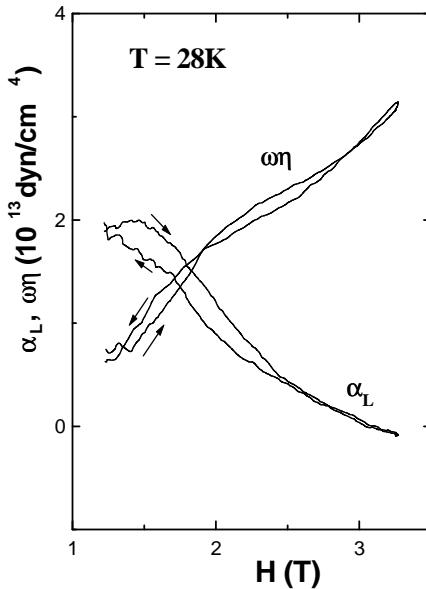


FIG. 4: Reconstruction of the coefficient of friction  $\omega\eta$  and Labusch parameter  $\alpha_L$ .

from our analysis:  $\sqrt{1 + \tan^2 \varphi_0}$  (Fig. 3b). Knowing  $\varphi_0$ , we can estimate the depth of the skin layer in the normal phase at the frequencies used here ( $\delta \approx 2.3 \times 10^{-3}$  cm) and the resistivity at  $T \geq T_c$  ( $\rho_n = 12.5 \mu\Omega \cdot \text{cm}$ ).

Relation (2) can easily be inverted to extract the real and imaginary parts of  $\alpha^*$  from experimental data without any additional assumptions. Of course, this can be done in the case when one is able to track the variation of both the modulus and phase of  $E_H$  in the accessible magnetic field region. It is also obvious that this procedure is doable only in that field region where the amplitude and phase of  $E_H$  deviate noticeably from their limiting values. The result of such a construction is shown in Fig. 4.

As expected,  $\eta$  varies practically linearly with applied field. For a known  $\sigma_n$ , the slope of this relation is determined, in accordance with Ref. 11, by the value of  $H_{c2}$ . Extrapolation of the data in Fig. 4 by a straight line passing through the origin gave  $H_{c2} \approx 3.2$  T, practically equal to the field that determines the beginning of the change in the phase of  $E_H$  at  $T = 28$  K (Fig. 3a). This means that in the MgB<sub>2</sub> sample under study,  $H_{c2}$  is limited by the condition of overlap of the cores of the vortices and not by the Clogston paramagnetic limit.[12]

It is seen from the results presented in Fig. 3 that the

main change in the phase of  $E_H$  is practically finished by the start of the deviation of  $|E_H|$  from the linear dependence characterizing the normal phase. This means that in this stage the inequality  $\omega\eta < \alpha_L \leq q^2 C_{44}$  holds, and  $\alpha_L$  can be estimated either by inverting relation (2) with  $\alpha^* \approx \alpha_L$  or simply from the relation  $\alpha_L \approx q^2 H_m^2 / 4\pi$ , where  $H_m$  is the characteristic field at which the transition of  $|E_H|$  from one linear trend to another occurs. In particular, for  $T = 20$  K an estimate of the Labusch parameter gives  $\alpha_L \sim 4 \times 10^{13}$  dyn/cm<sup>4</sup>. Using the order-of-magnitude relation  $\alpha_L \approx BI_c/cr$ , where  $I_c$  is the critical current and  $r$  is the characteristic spatial scale of the pinning potential, which for  $H \gg H_{c1}$  is close to the period of the vortex structure, we obtain  $I_c \approx 3 \times 10^4$  A/cm<sup>2</sup> ( $T = 20$  K,  $H \approx 3$  T).

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